

The University of Chicago

HIGH CENTRIFUGAL FIELDS AND RADIOACTIVE DECAY

A PART OF A DISSERTATION SUBMITTED TO
THE FACULTY OF THE DIVISION OF THE
PHYSICAL SCIENCES IN CANDIDACY FOR
THE DEGREE OF DOCTOR OF PHILOSOPHY

DEPARTMENT OF CHEMISTRY

1941

By

ARTHUR H. JAFFEY

Reprinted from
THE PHYSICAL REVIEW
Vol. 63, January 1 and 15, 1943

The University of Chicago

HIGH CENTRIFUGAL FIELDS AND RADIOACTIVE DECAY

A PART OF A DISSERTATION SUBMITTED TO
THE FACULTY OF THE DIVISION OF THE
PHYSICAL SCIENCES IN CANDIDACY FOR
THE DEGREE OF DOCTOR OF PHILOSOPHY

DEPARTMENT OF CHEMISTRY

1941

By
ARTHUR H. JAFFEY

Reprinted from
THE PHYSICAL REVIEW
Vol. 63, January 1 and 15, 1943



High Centrifugal Fields and Radioactive Decay

A. H. JAFFEY

George Herbert Jones Laboratory, University of Chicago, Chicago, Illinois

(Received October 6, 1942)

Previous experiments to detect some influence on the decay process of a naturally radioactive substance by means of centrifugal fields (20,000 g) had yielded no effect within 0.1 percent. It seemed of value to re-examine such a possibility in view of the high centrifugal fields (order of 1,000,000 g) now available, the new types of decay processes in artificial radioactivity, and the non-electromagnetic forces now recognized as operating in atomic nuclei. Except for fission an example of each type of radioactive decay was investigated. Two methods were em-

ployed, one of which made use of a toroidal tube counter surrounding the spinning rotor. An ionization chamber furnished a precision of several tenths of a percent and the counter allowed a precision of several percent. No definite effect was found. However, in Br^{80} where a transition occurs between isomers by emission of gamma-rays rather than the usual transformation within the nucleus involving charged particles, there was some sign of systematic deviation from the accepted half-lives in a centrifugal field of 632,000 g but it may be within the total experimental error.

MANY experiments, very diverse in character, have all failed to effect any measurable change in the decay processes of naturally radioactive substances.¹ The discovery of artificial

radioactivity throughout the periodic system, the appearance of new types of nuclear transformations and the new basic particles, the neutron and the positron, have brought to the fore the need of attempting once again to influence the decay processes and thus to link

¹F. W. Kohlrausch, "Radioaktivität," *Handbuch der Experimental Physik* (Leipzig, 1928), Vol. 15, p. 669.

these phenomena with the main body of physics and chemistry.

Such experiments now seem especially timely because of the failure of electromagnetic theory to account for results in scattering experiments where the interactions of proton-proton and neutron-proton (particles unlike in charge but of about the same mass)² are of the same magnitude. Hence non-electromagnetic forces, still very obscure, are now entertained as operative in the structure and properties of atomic nuclei. Also not clear is the importance of the theory of relativity in intra-nuclear dynamics although extreme concentration of matter and intensity of the fields are accepted as present in nuclei. Because of the equivalence of gravitational and accelerational fields and the availability of very high centrifugal fields these investigations were undertaken to determine what influence fields of about 500,000 g to 1,000,000 g have on the radioactive intensity at a given instant and on the rate of its decay in substances which represent every radioactive decay process now known except fission.

Rutherford and Compton, and Compton³ had found that centrifugal fields about 20,000 g did not affect the intensity of the gamma-radiation from a radon bulb by as much as 0.1 percent. Effects due to centrifugal fields are hardly to be expected on grounds of energy alone since the potential energy due to rotation, in the present experiments, is only of the order of 0.1 ev whereas nuclear transformations are attended by energy changes of the order of 10^6 ev. What was sought was a response to a particular kind of influence rather than any effect ascribable solely to a

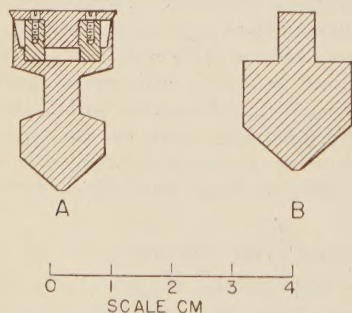


FIG. 1. Rotors.

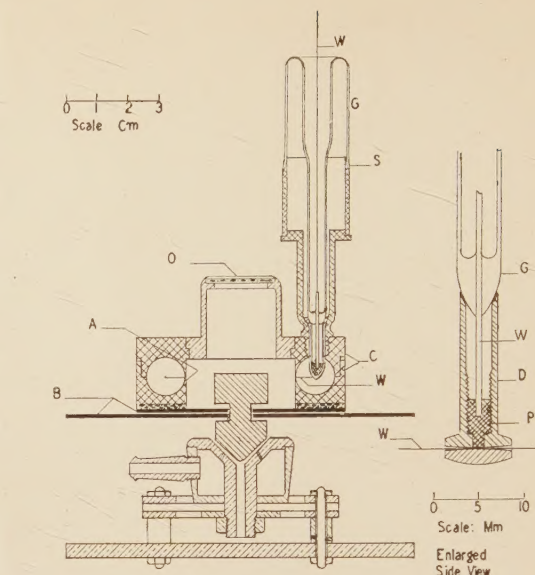


FIG. 2. Toroidal counter with rotor on stator. W—tungsten wire; A—copper foil window; B—baffles; C—copper case; D—lavite insulator; G—glass; O—glass window for observing rotation with stroboscope; P—copper plug; S—glass-copper seal.

change in potential energy. Moreover, what are involved are not changes in equilibrium but in rate processes. Rates of many chemical reactions are greatly affected by small net energy changes of the proper kind. On the basis of energy it would be preferable to investigate critical or resonance situations as with thermal neutrons.

The experiments performed divide into two sets. The first in which a Geiger-Mueller tube counter was employed and in the second, an ionization chamber and amplifier. The more sensitive method (set I) had been found necessary because the only means available at the time for inducing radioactivity was a weak neutron source (200 mC radon-beryllium). The two sets differ also in the method for detecting whether the centrifugal field had effected any change in the activity. In set I, the half-lives of the disintegration from the substances in rotation were compared with the published values.⁴

⁴ This comparison was feasible when the decay could be adequately represented by one exponential term. The following procedure was adopted when the counting rate is given as the sum $N = \sum_i N_{0i} e^{-\lambda_i t}$ over i , the separate periods contributing to the decay. The value of the initial intensity N_0 for each period was obtained from the experimental data by use of the method of least squares and by assuming the published values for λ_i to be correct. In order to avoid the use of weighting factors, equal numbers

² M. A. Tuve, N. P. Heydenberg, and L. R. Hafstad, Phys. Rev. **50**, 806 (1936).

³ E. Rutherford and A. H. Compton, Nature **104**, 412 (1919); A. H. Compton, Phil. Mag. **39**, 659 (1920).

In set II, instead of comparing the half-lives, the actual radioactive intensities for high speed and low speed were compared by means of the corresponding currents in an ionization chamber. It may be readily shown⁵ that to a first approxi-

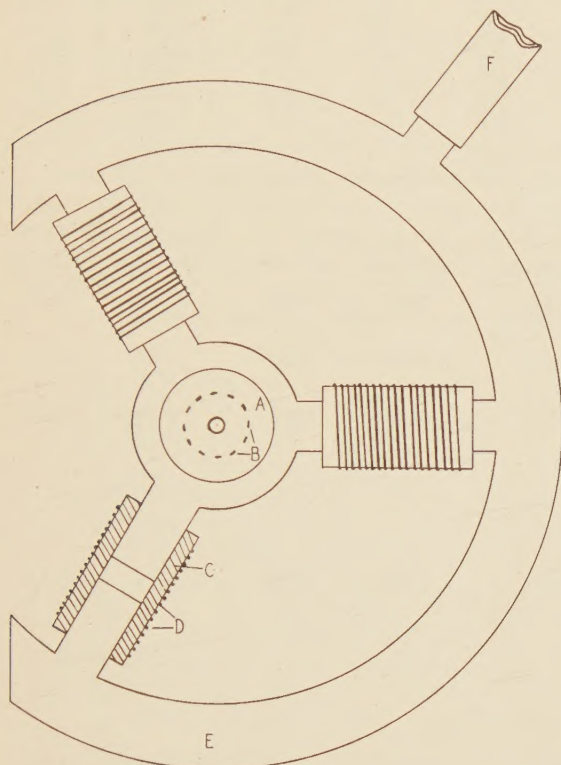


FIG. 3. Symmetrical stator mount. One air inlet shown in cross section. A—chromium plated cone; B—air jets; C—rubber air inlet; D—tightly wound and soldered copper wire; E—hollow air distributor; F—rubber hose from air valve and gauge.

of counts were taken for each experimental point thus making the relative probable error for that point approximately equal to the errors of any other point. If any systematic deviation between the curve calculated from the N_0 and λ_i and the experimental points is appreciably larger than the experimental error, it may be concluded that at least one of the decay constants is significantly different from the assumed value.

⁵ Assume the exponential decay law, $N = N_0 e^{-\lambda t}$ then $dN/dt = -\lambda N_0 e^{-\lambda t} = -\lambda N$. If Δq is the charge collected in the ionization chamber in the time interval Δt , and if the relative positions of the radioactive material and the ionization chamber remain unchanged, then Δq is proportional to ΔN , the number of particles disintegrating during Δt . If V is the measured voltage and i the current through the grid resistor, $V = i R_{\text{grid}} = (dq/dt) R_{\text{grid}} \propto (dN/dt)$ so $V \propto \lambda N$. If the speed of the rotor can be changed rapidly enough so that N , the number of radioactive atoms present, remains practically constant, then a change in the decay constant with speed will be reflected in a change in the measured voltage. If the decay is too rapid for direct comparison, i.e., if during the interval required to reach the final value in speed the value of N changes appreciably, then a modification of the procedure is necessary. Measurements are taken at two values of the rotational speed, the

mation any change in the half-lives is proportional to a change in ionization current. This method is not restricted to a single decaying species and requires none of the computational fitting of the first method for groups of atoms disintegrating at the same time, since it is about as simple to determine the displacement of a complex decay curve as of a decay which follows a straight line.

EXPERIMENTAL

The rotors⁶ were air driven and supported (Fig. 1A and 1B for rotors, and for stators, Fig. 3 and lower portion of Fig. 2). Where possible the material to be activated constituted a part of the cylindrical surface of the rotor, usually as electroplated metal. The rotors were machined out of beryllium-copper, an alloy of high tensile strength. During activation all but the significant cylindrical surface was heavily shielded. When the substance did not lend itself to this procedure, the activation was performed on the material in powder form and if necessary it was dissolved and co-precipitated with an appropriate substance. The active powder was placed in the thin-walled cavity of the rotor (Fig. 1A).

The rotor represented in Fig. 1A owed its irregular shape to the need of baffles to keep the driving air from impinging on the window of the counter and on objects near it. It had been found⁷ that the air deposited the decay products of radon and thoron from the atmosphere on almost all materials exposed to the air stream and depending on the weather, the counts due to this source may reach as many as fifty per minute. Even with the precaution of baffles, the deposited activity often amounted to as much as ten counts

highest attainable with the air pressure available and the lowest at which the rotors will spin stably. The voltage is measured at short intervals for a number of readings at one of the speeds and the speed is changed to the other after each reading. The resulting voltage (or log voltage) is plotted against time. If an effect of the kind sought is present, the points taken at high speed should lie on a curve displaced somewhat from the curve passing through the points taken at low speed; if the effect is absent all the points should lie on the same smooth curve. It has been assumed that the form of the decay law does not change when the radioactive substance is placed into an accelerational field and that any accelerational effect is expressible simply as a change in λ , an assumption valid in the first approximation for any small effects.

⁶ E. Henriot and E. Huguenard, *Comptes rendus* **180**, 1389 (1925), *J. de phys. et rad.* [6] **8**, 433 (1927). J. W. Beams and E. G. Pickels, *Rev. Sci. Inst.* **6**, 299 (1935).

⁷ S. Freed and M. L. Schultz, *J. Frank. Inst.* **231**, 345 (1941).

TABLE I. Decay in centrifugal fields of substances showing single decay periods. Method I of text.

Radioactive isotope	Literature half-life	Observed half-life	Air effect counts/min.	Initial intensity counts/min.	Final intensity counts/min.	R cm	n rev./sec.	Centrifugal field $g \times 10^{-5}$
Mn ⁵⁶	2.59 $\pm$ 0.02 hr.	2.65 $\pm$ 0.03 hr.	0	172	29	0.79	4025	5.16
Mn ⁵⁶	2.59 $\pm$ 0.02 hr.	2.55 $\pm$ 0.08 hr.	0	60	17	0.85	4000	5.48
Cl ³⁸	37.40 min.	39.1 $\pm$ 0.2 min.	15	1489	43	0.79	4025	5.16
Cu ⁶⁴	12.8 $\pm$ 0.1 hr.	13.01 $\pm$ 0.02 hr.	5	1420	224	0.85	3700	4.69
ThC'	60.8 min.	62.4 $\pm$ 0.4 min.	0	614	17	0.79	4000	5.09
ThC'	60.8 min.	58.0 $\pm$ 0.6 min.	7	353	18	0.79	3950	4.97
ThC'	60.8 min.	59.2 $\pm$ 1.0 min.	1	143	9	0.79	3950	4.97
ThC'	60.8 min.	61.2 $\pm$ 1.6 min.	9	316	126	0.79	3900	4.84

TABLE II. Decay in centrifugal fields of substances possessing several radioactive periods. Method II of text.

Metal activated	Radioactive isotopes present	Half-life (literature)	Type of radiation	Precision*	Rev. per sec.		Centrifugal field† $g \times 10^{-5}$	
					Maximum	Minimum	Maximum	Minimum
Cu	Cu ⁶⁴	12.8 hr.	β^- β^+ and K	0.25%	4000	3150	5.63	3.49
Cu	Cu ⁶⁴	12.8 hr.	β^- β^+ and K	0.20%	3700	2700	4.82	2.56
Cd	Cd ¹¹⁷	3.75 hr.	β^-	0.10%	5250	2850	9.70	2.86
	In ¹¹⁷	117 min.	β^-					
	Cd ¹¹⁵	2.5 days	β^- and γ					
	In ^{115*}	4.1 hr.	I.T., γ and internally converted electrons					
Zn	Zn ⁶⁹	57 min.	β^-	0.20%	5100	2675	9.15	2.52
	Ga ⁶⁸	68 min.	β^+					
	Zn ⁶⁹	13.8 hr.	I.T., γ					
	Ga ⁶⁷	83 hr.	K , γ and internally converted electrons					
Ni	Cu ⁶¹	3.4 hr.	β^+ , K	0.07%	5125	2025	9.24	1.44
	Cu ⁶⁶	5 min.	β^-					
	Ni ⁶³	2.6 hr.	β^- , γ					
Cr	Mn ⁵¹	46 min.	β^+	0.20%	5050	2575	8.97	2.33

* Represents the average absolute deviation of the observed points from the smooth curve drawn through them.

† The radius of each rotor was 0.872 cm.

per minute and had to be determined with unactivated rotors and taken into account (Table I, column 4). The strong radioactivity obtained with deuterons from the cyclotron of this university (set II) made it possible to ignore the air effect and hence a rotor of simple form could be used, which attained about 1,000,000 g (radius = 0.872 cm).

The feeble radioactivity of set I called for a counter so shaped as to surround the rotor. Figure 3 shows its approximate toroidal form. The copper window A , about 0.05 mm thick, was soldered with tin to the copper case B . The latter was made of two rings of copper into each of which was machined a semitoroidal groove so as to form a toroidal cavity within the block M

when the two halves were screwed together. The cylindrical copper "window" was co-axial with the rotor. The counter possessed a plateau from 300 volts to 400 volts.⁸

⁸ It has been treated by the method of O. S. Duffendack, A. Lifschutz, and M. M. Slawsky [Phys. Rev. **52**, 1231 (1937)]. The circuit of H. V. Neher and W. H. Pickering [Phys. Rev. **53**, 317 (1938)] served to quench the counter discharge and also maintained the counter case at ground potential. A twofold scaling circuit was employed similar to the first stage of the scale of eight of W. G. Shepherd and R. D. Haxby [Rev. Sci. Inst. **1**, 425 (1936)]. The counter was corrected for non-linearity, i.e., the losses due to the finite resolving time of the circuit and of the mechanical recorder according to the procedure of Duffendack, Lifschutz, and Slawsky and of Lifschutz and Duffendack [Phys. Rev. **54**, 714 (1938)]. The basic count varied from 10 to 30 per minute depending mostly on the size of the counter. The method of evaluating the decay rates from the data followed H. Peierls [Proc. Roy. Soc. **A149**, 467 (1935)].

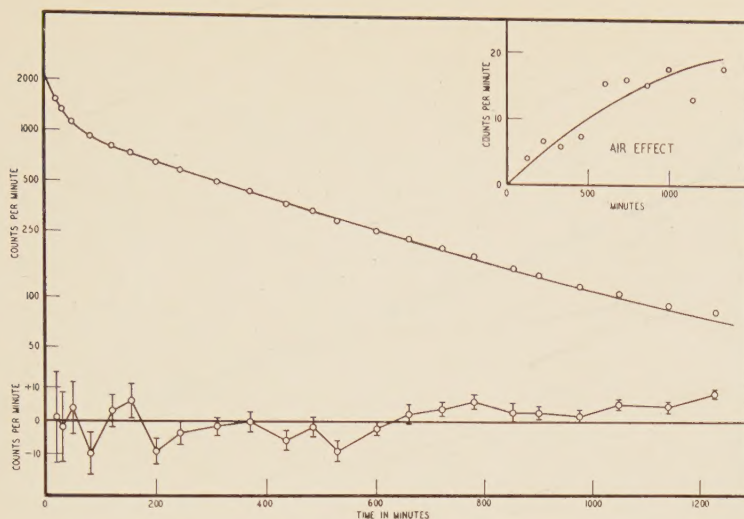


FIG. 4. Decay of activated bromine.

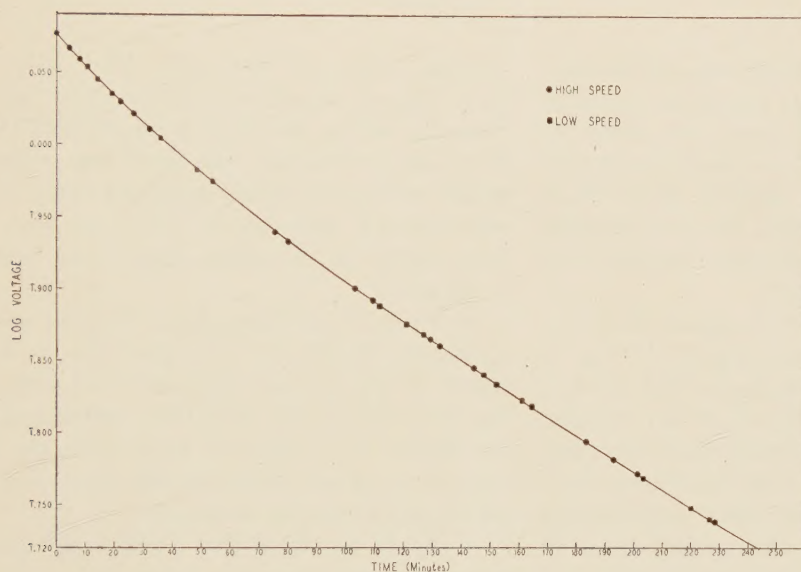


FIG. 5. Decay of activated cadmium.

MEASUREMENTS

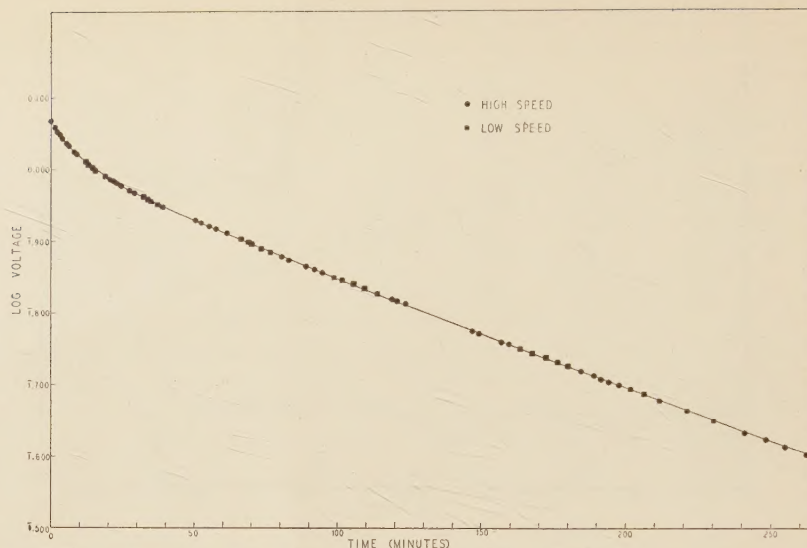
The first set of measurements, those listed in Table I and in addition those on aluminum and bromine, both of which decay with several periods, revealed no definite change in the radioactive decay in a centrifugal field within an estimated 3 percent. The actual half-life obtained from copper exceeds by more than this the published values which, moreover, are badly scattered. Separate experiments carried out in connection with set II (see Table II) revealed that the higher value arises from the presence of Zn^{65} with half-life 250 days formed during the activation. The method (as employed in set II) of

detecting any change which the centrifugal field might bring about, established that the decay rate remained undisturbed within 0.25 percent when the centrifugal field changed from $2.5 \times 10^5 \text{ g}$ to $5.6 \times 10^5 \text{ g}$. The discrepancy in half-life as given in Table I may then be regarded as removed.

The decay of Cu^{64} is of especial interest. It decays by β -ray emission to form Zn^{64} and by positron emission or K -electron capture to form Ni^{64} . Both transformations proceed at the same rate.⁹ Under the influence of the centrifugal field some readjustment might occur in the relative intensities of the β -ray and positron activities.

⁹ S. N. Van Voorhis, Phys. Rev. 50, 895 (1936).

FIG. 6. Decay of activated nickel.



With bromine the possibility existed of influencing a process with no net change in charge in the nucleus, the transition from one isomer of Br^{80} to another. Br^{80} has been shown to have a 4.5-hr. period due to an excited state which decays by gamma-radiation to the ground state, which in turn decays by electron emission with an 18.5-min. period¹⁰ to Kr^{80} .

The decay curve for Br^{80} in which the 33-hr. period is taken into account is given in Fig. 4. The experimental values are shown with vertical lines. An examination of the plot, on an enlarged scale, indicates a small systematic trend (starting at about 200 min.) between the calculated curve and the experimental values. Since the standard deviations represented by the vertical lines do not include the errors in correction for the non-linearity and air-effects, this trend may not be outside the total experimental error since, as is shown in the inset, the air-effect in these experiments was unusually large.

A special procedure was devised for bromine. Lithium bromide as powder was activated in a beam of 8.5-Mev deuterons, then dissolved in water and silver nitrate added. The precipitated silver bromide was fused and the molten salt was transferred to the silver-plated cylindrical surface of a rotor whose temperature was just above the

melting point of the salt. The silver surface insured excellent wetting and adhesion; chemical action also had been avoided. Before being set in rotation, the layer was vigorously buffed to remove loosely adhering material. The salt adhered perfectly at 4975 rev./sec. on a rotor 0.834 cm in radius (centrifugal field 6.32×10^5 g).

In Table II are listed data obtained with the ionization chamber.¹¹ The precision is several tenths of one percent. Figures 5 and 6 illustrate the precision of the data. The centrifugal fields are higher than those of set I since the rotor, because of its simpler form and reduced surface, encountered less air resistance.

It should be added that Table I also includes several measurements on naturally radioactive thorium C' isolated in different ways.

We wish to express here our gratitude to the group working with the cyclotron of this university and especially to Dr. Louis Slotin.

¹⁰ A. H. Snell, Phys. Rev. **52**, 1007 (1937); E. Segrè, R.S. Halford, and G. T. Seaborg, Phys. Rev. **55**, 321 (1939); D. C. DeVault and W. F. Libby, Phys. Rev. **55**, 322 (1939).

¹¹ The design of the ionization chamber was similar to that of S. W. Barnes [Rev. Sci. Inst. **10**, 1 (1939)] and the current was amplified with an FP-54 electrometer tube. The amplifier circuit was essentially that described by D. B. Penick [Rev. Sci. Inst. **6**, 115 (1935)]. To increase the range and precision it was converted into a null instrument by balancing the ionization current with an accurate potentiometer in the grid circuit. The beam of deuterons was usually directed at the material on the rotor within the vacuum chamber of the cyclotron. The rotor with its shielding of massive copper was placed on a water-cooled spindle entering the vacuum chamber through a Wilson seal [Rev. Sci. Inst. **12**, 91 (1941)]. So disposed, the rotor could be turned in the vacuum at intervals to achieve a rather uniform activation over the surface.

